Supporting Information for

Dense Arrangement of Crown Ethers in Graphene: Novel Graphitic Carbon Oxides with Enhanced Optoelectronic Properties

HongYan Li[a], Jiang Xiang[a], Liang Chen[b,a], Jing Xu[a,] and Wei Liu [a,*]*

[a] Department of Optical Engineering, College of Optical, Mechanical and Electrical Engineering, Zhejiang A&F University, Hangzhou, Zhejiang, 311300, P. R. China

[b] School of Physical Science and Technology, Ningbo University, Ningbo, Zhejiang, 315211, P. R. China

Email: weiliu@zafu.edu.cn; jingxu@zafu.edu.cn.

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Table S1. The nanopore density of CmG-n (m=3-6, n=1-6).

System	C_xO_y	primitive cell area ($Å^2$)	nanopore density ($Å^{-2}$)	System	C_xO_y	primitive cell area ($Å^2$)	nanopore density ($Å^{-2}$)
C3G-1	C_4O_3	20.22	4.95	C5G-1	$C_{28}O_{20}$	144.59	2.77
C3G-2	$C_{12}O_6$	51.24	3.90	C5G-2	$C_{14}O_{10}$	73.24	2.73
C3G-3	$C_{16}O_6$	60.59	3.30	C5G-3	$C_{20}O_{10}$	90.82	2.20
C3G-4	$C_{16}O_6$	60.72	3.29	C5G-4	$C_{10}O_5$	45.46	2.20
C3G-5	C_8O_3	30.93	3.23	C5G-5	$C_{10}O_5$	45.58	2.19
C3G-6	$C_{68}O_3$	189.21	0.53	C5G-6	$C_{64}O_5$	188.27	0.53
C4G-1	C_6O_4	28.73	3.48	C6G-1	C_6O_6	39.16	2.55
C4G-2	C_8O_4	35.64	2.81	C6G-2	$C_{12}O_6$	58.70	1.70
C4G-3	$C_{36}O_{16}$	146.00	2.74	C6G-3	$C_{12}O_6$	58.93	1.70
C4G-4	$C_{18}O_8$	75.13	2.66	C6G-4	$C_{12}O_6$	59.24	1.69
C4G-5	$C_{18}O_8$	76.22	2.62	C6G-5	$C_{14}O_6$	64.70	1.55
C4G-6	$C_{66}O_4$	188.60	0.53	C6G-6	$C_{60}O_6$	186.55	0.54

a.

b.

c.

C3G-2 (C$_{12}$O$_6$)

C3G-3 (C$_{16}$O$_6$)

C3G-4 (C$_{16}$O$_6$)

d.

e.

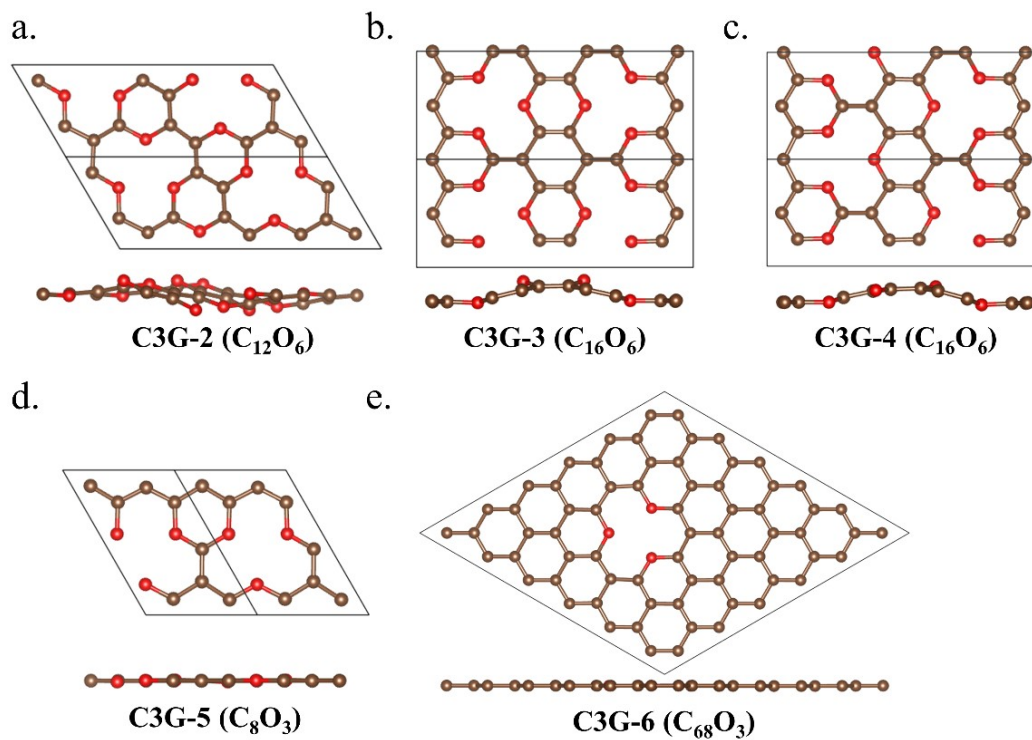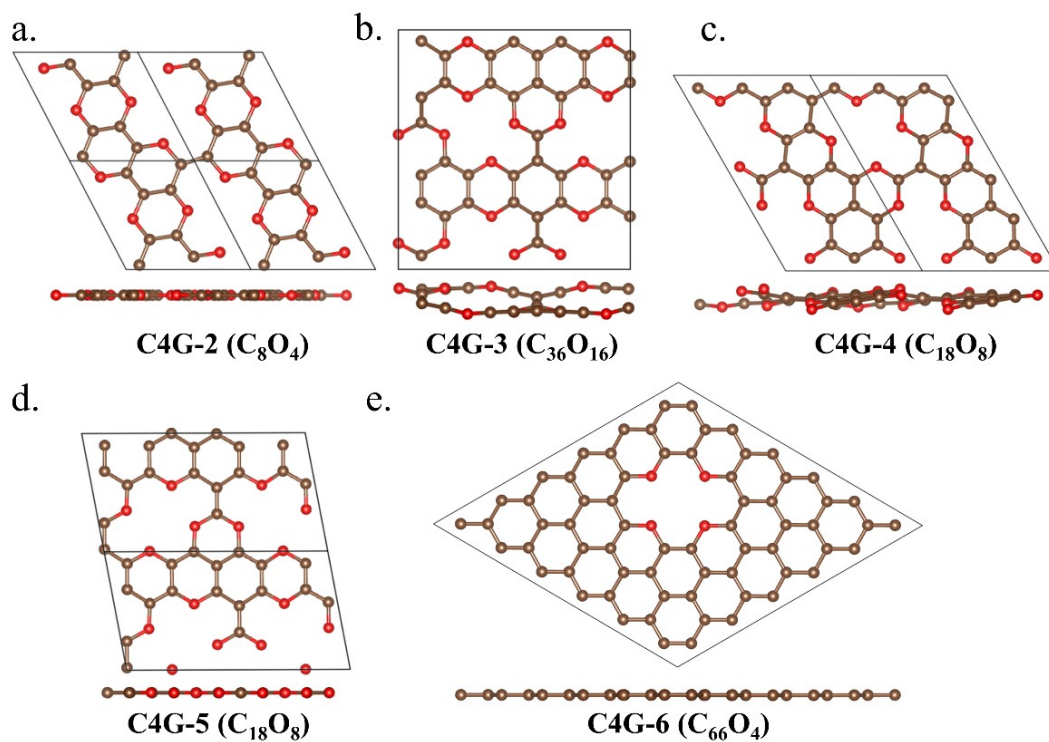**C3G-5 (C$_8$O$_3$)**

C3G-6 (C$_{68}$O$_3$)

Figure S1. Top and side views of the optimized structures of (a) C3G-2, (b) C3G-3, (c) C3G-4, (d) C3G-5 and C3G-6.

Figure S2. Top and side views of the optimized structures of (a) C4G-2, (b) C4G-3, (c) C4G-4, (d) C4G-5 and C4G-6.
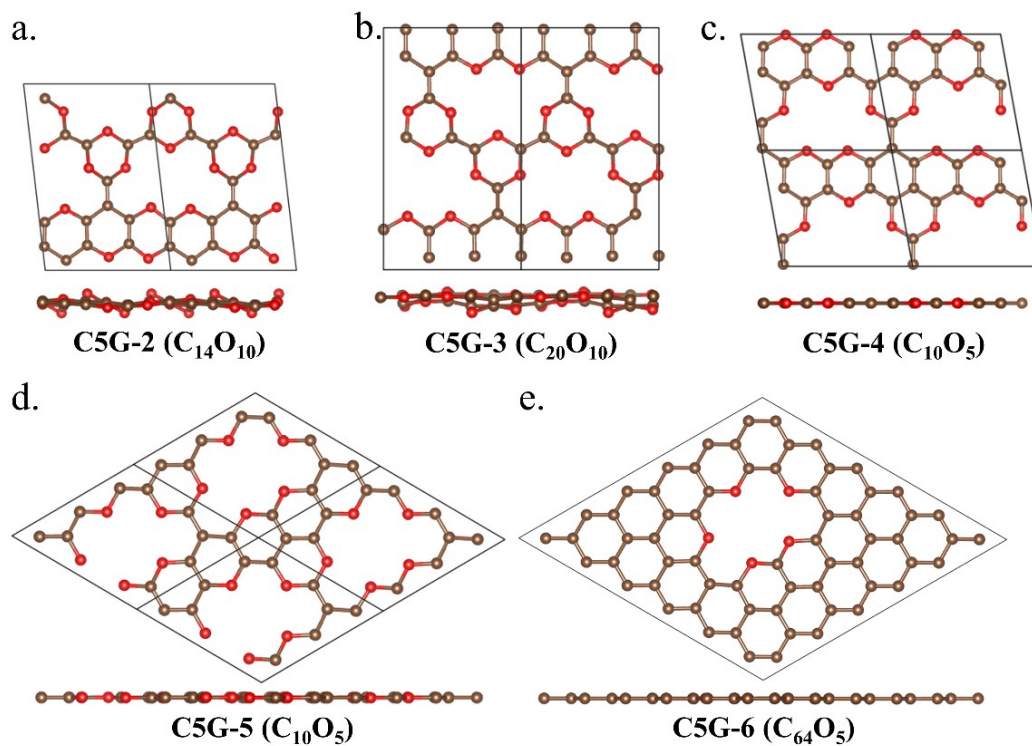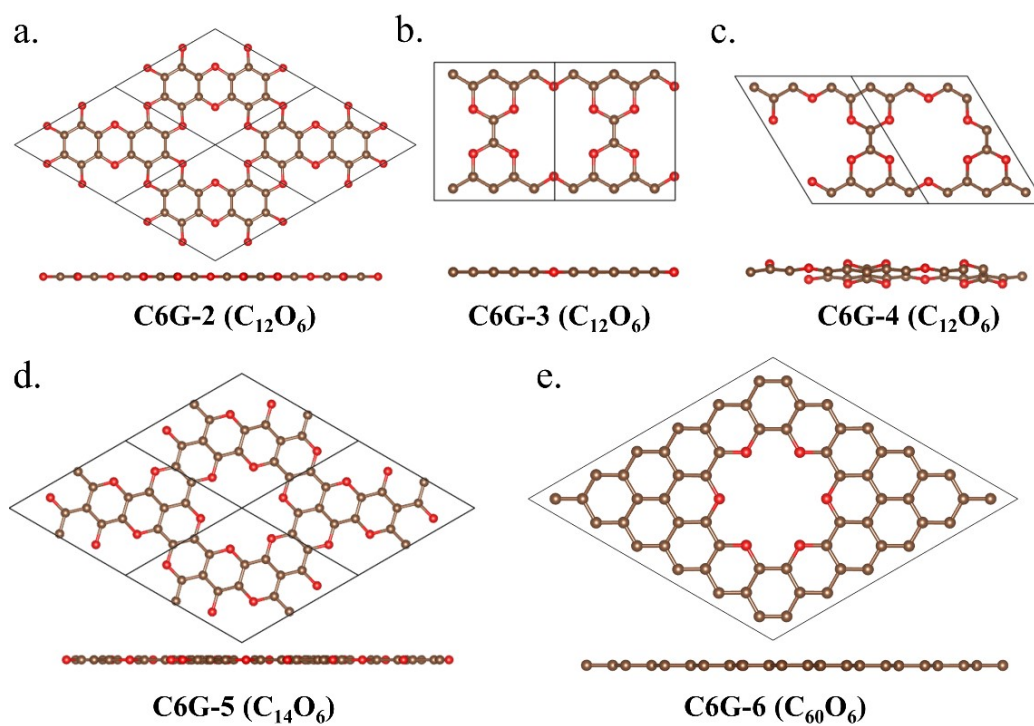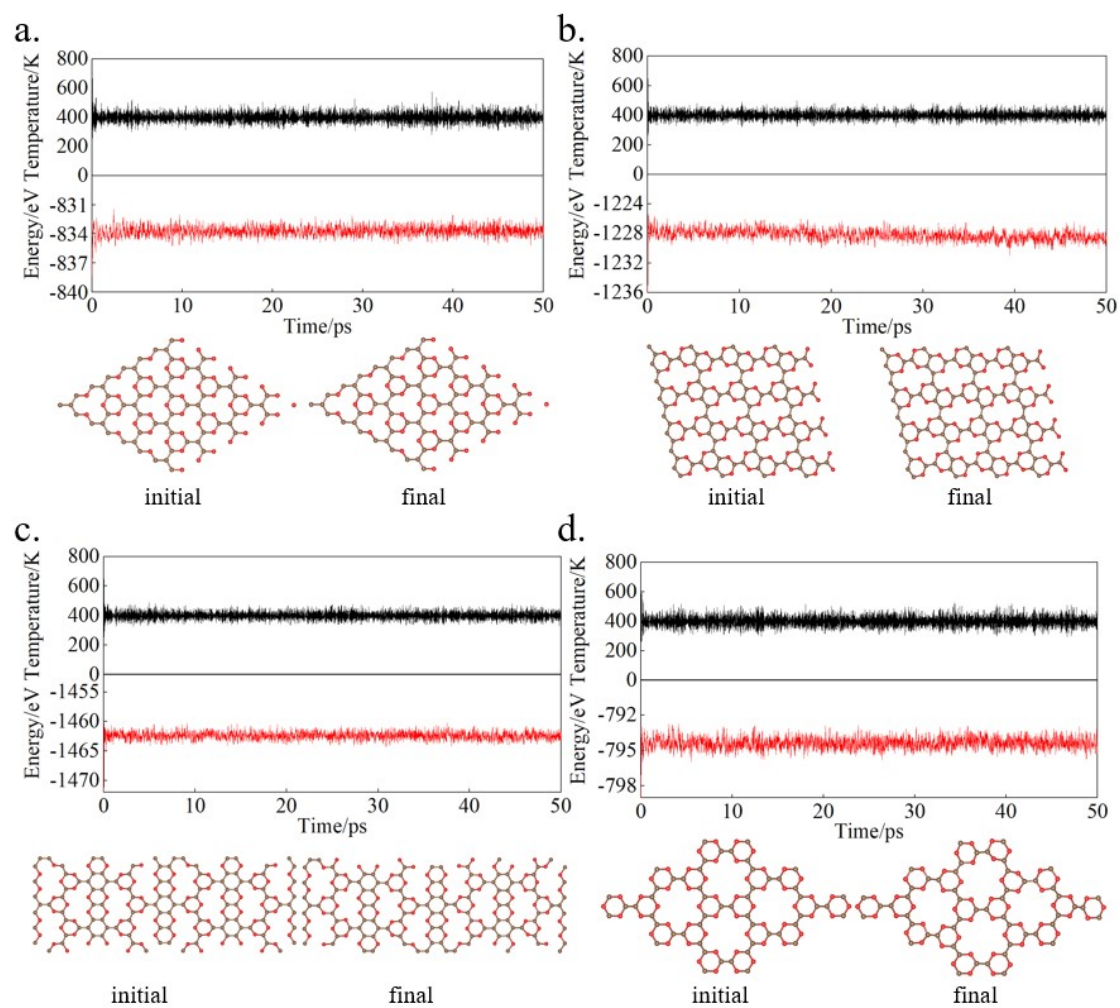
a.

b.

c.

C5G-2 (C$_{14}$O$_{10}$)

C5G-3 (C$_{20}$O$_{10}$)

C5G-4 (C$_{10}$O$_{5}$)

d.

e.

C5G-5 (C$_{10}$O$_{5}$)

C5G-6 (C$_{64}$O$_{5}$)

Figure S3. Top and side views of the optimized structures of (a) C5G-2, (b) C5G-3, (c) C5G-4, (d) C5G-5 and C5G-6.

a.

b.

c.

C6G-2 (C$_{12}$O$_6$)

C6G-3 (C$_{12}$O$_6$)

C6G-4 (C$_{12}$O$_6$)

d.

e.

C6G-5 (C$_{14}$O$_6$)

C6G-6 (C$_{60}$O$_6$)

Figure S4. Top and side views of the optimized structures of (a) C6G-2, (b) C6G-3, (c) C6G-4, (d) C6G-5 and C6G-6.

Figure S5. Fluctuation of temperature and energy against time for AIMD simulations at 400K: (a) C3G-1, (b) C4G-1, (c) C5G-1, (d) C6G-1. The initial and final snapshots of the simulations are also shown.
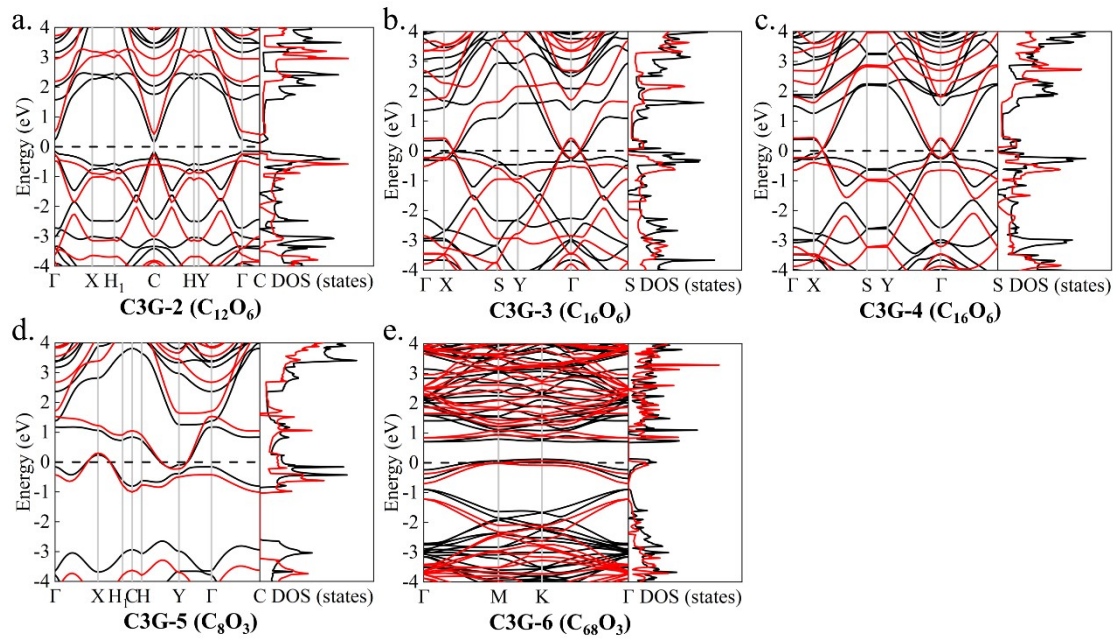
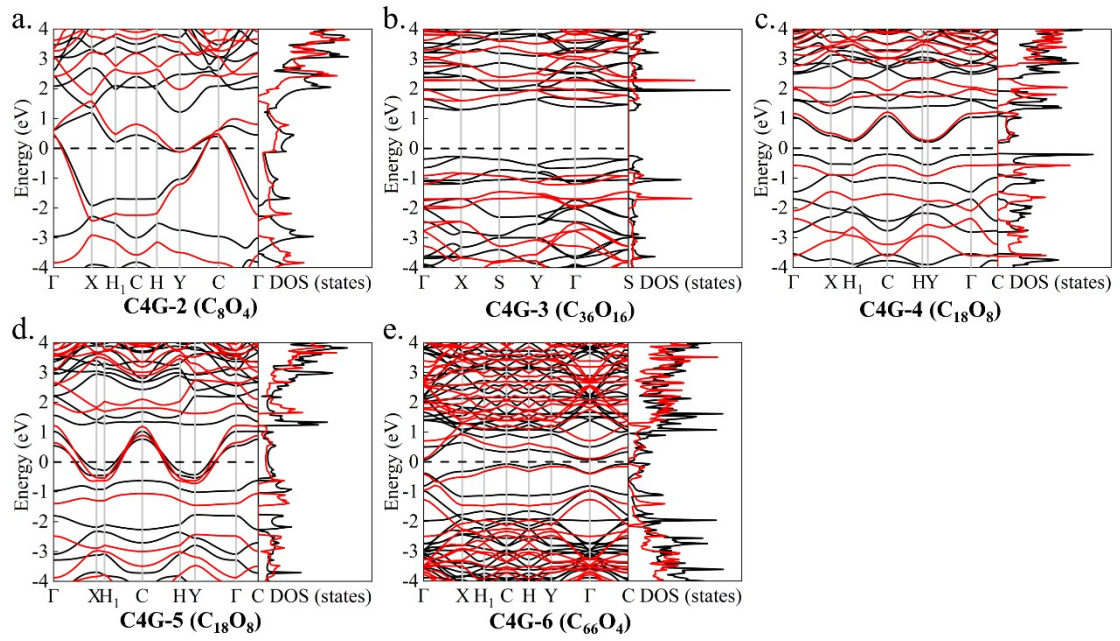**Figure S6.** The band structures and DOS of (a) C3G-2, (b) C3G-3, (c) C3G-4, (d) C3G-5 and (e) C3G-6.

Figure S7. The band structures and DOS of (a) C4G-2, (b) C4G-3, (c) C4G-4, (d) C4G-5 and (e) C4G-6.
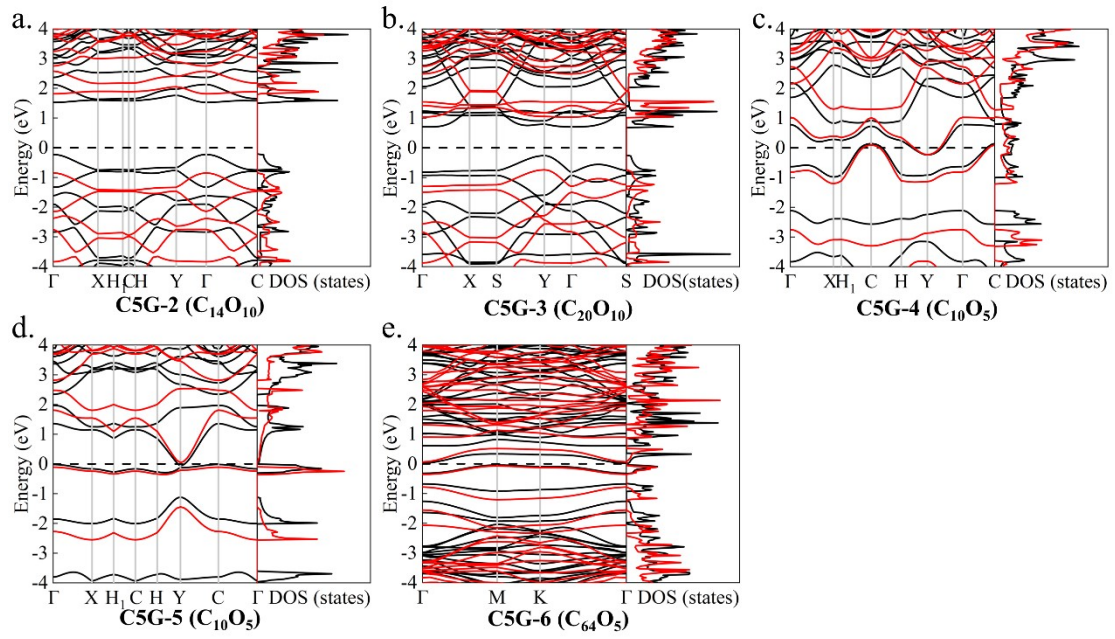
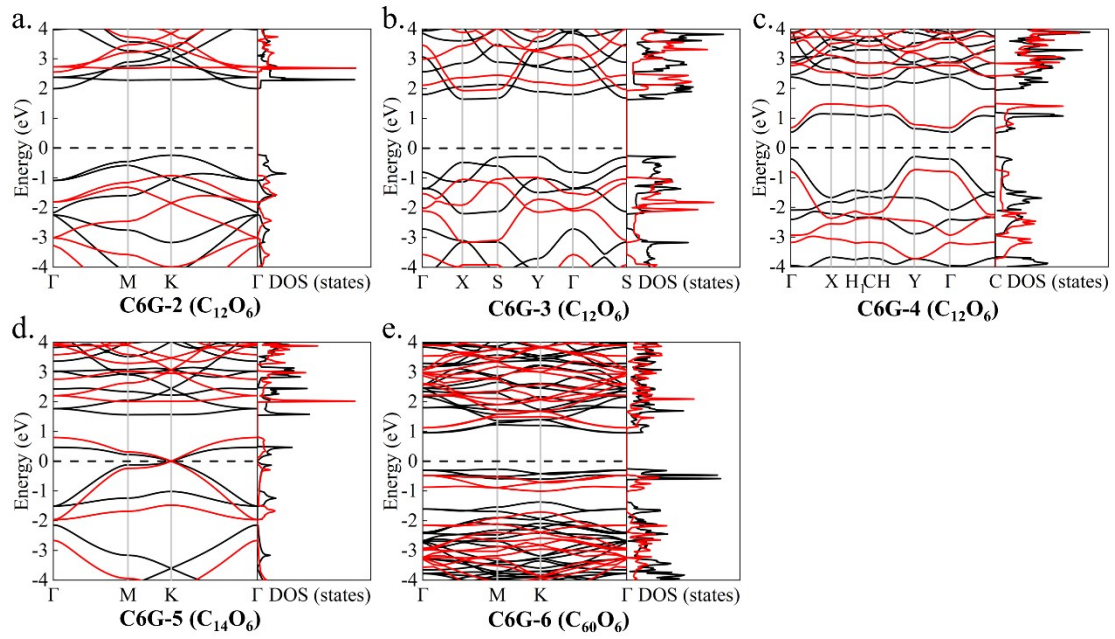**Figure S8.** The band structures and DOS of (a) C5G-2, (b) C5G-3, (c) C5G-4, (d) C5G-5 and (e) C5G-6.

Figure S9. The band structures and DOS of (a) C6G-2, (b) C6G-3, (c) C6G-4, (d) C6G-5 and (e) C6G-6.
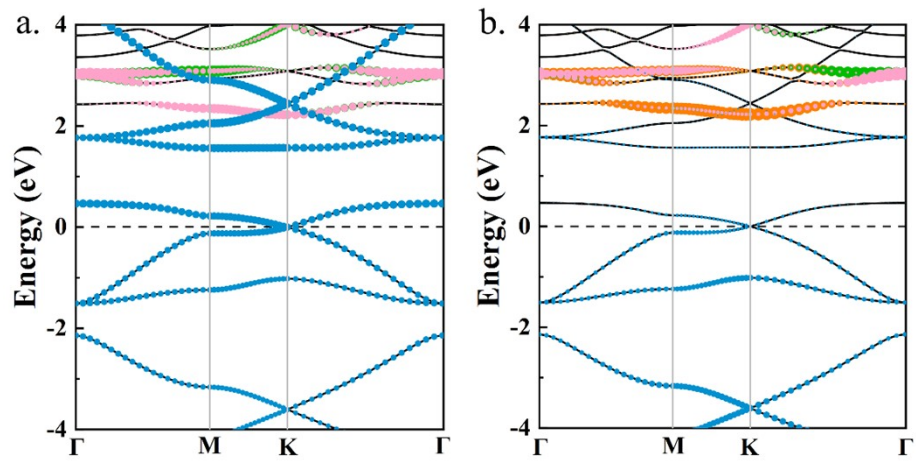
Figure S10. Orbital-projected band structures of (a) C atoms and (b) O atoms in C6G-5. The orange, green, pink and blue points represent the s, p_x, p_y and p_z orbitals of C/O, respectively.
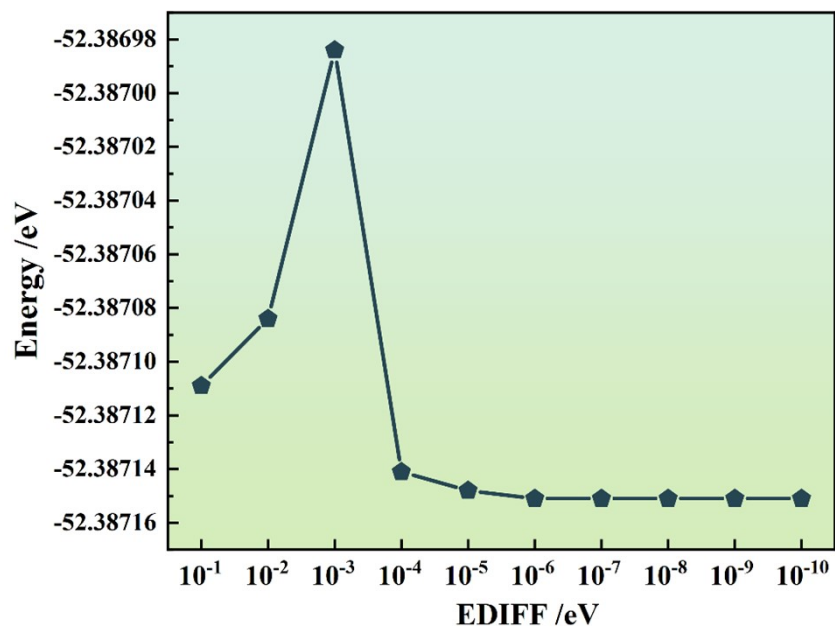
Figure S11. A diagram of the energy convergence standard test.